(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 688 773 B1

(12) EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention of the grant of the patent:
20.05.1998 Bulletin 1998/21

(51) Int Cl.[6]: **C07D 471/04**, A01N 43/90
// (C07D471/04, 235:00, 221:00)

(21) Application number: **95810410.1**

(22) Date of filing: **20.06.1995**

(54) 2-Phenyl-7-chloro-perhydroimidazo(1,5-a)pyridines

2-Phenyl-7-chlor-perhydroimidazo(1,5-a)pyridine

2-Phényl-7-chloro-perhydroimidazo(1,5-a)pyridines

(84) Designated Contracting States:
CH DE FR GB IT LI

(30) Priority: **23.06.1994 GB 9412603**

(43) Date of publication of application:
27.12.1995 Bulletin 1995/52

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(56) References cited:
EP-A- 0 384 973
EP-A- 0 468 924
EP-A- 0 493 323

Remarks:

The file contains technical information submitted after the application was filed and not included in this specification

EP 0 688 773 B1

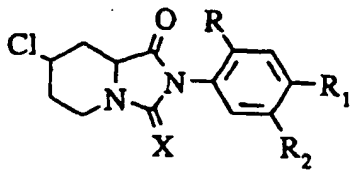
Printed by Jouve, 75001 PARIS (FR)

Description

The present invention relates to novel 2-phenyl-7-chloro-perhydroimidazo[1,5a]pyridines, synthesis thereof, and the use of said compound for controlling undesired weeds.

Herbicidal 7-fluoro-perhydroimidazopyridines are known from EP-A493 323 as effective compounds for combatting weeds. In practice however these compounds do not always satisfy the needs of selective weed control under all aspects, e.g. with respect to desired selectivity in crops of cultivated plants.

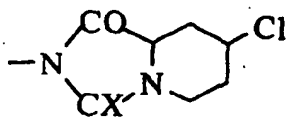
It has now been found that the 2-phenyl-7-chloro2-phenyl-7-chloro-perhydroimidazo[1,5a]pyridines of formula I



(I)

wherein

X	is oxygen or sulfur,
R	is hydrogen, chlorine or fluorine,
R_1	is fluorine, chlorine, bromine, cyano or methyl,
R_2	is halogen, C_{1-6}alkyl, C_{1-6}alkoxy, C_{1-6}alkylcarbonyloxy, C_{3-6}cycloalkoxy, C_{3-6}alkynyloxy, C_{3-6}alkenyloxy, -COOH, -CO-R_3, -CO-NR_4R_5, -NR_6R_7, C_{2-6}alkenyl, N-pyrrolyl, 2-oxo-3-tetrahydrofuranyl or a group



, or is
C_{1-6}alkyl substituted with halogen, COOH, -CO-R_3, -CO-NR_8R_9, =N-OH, =N-C_{1-4}alkoxy, -O-CO-R_3 or -O-C_{2-3}alkylene-O-; or is
C_{2-6}alkenyl substituted with halogen, COOH, -CO-R_3, -CO-NR_8R_9 or C_{1-4}alkoxy; or is
C_{1-6}alkoxy substituted with cyano, -CO-R_3, C_{1-4}alkoxy, C_{3-6}cycloalkyl, oxiranyl, or optionally substituted thienyl or phenyl with the substituents selected from C_{1-4}alkyl, C_{1-4}alkoxy or halogen; or is
C_{3-6}alkenyloxy substituted by C_{2-5}alkynyl; or

R_1 and R_2	together form a bridge member selected from -Y-CHR_{10}-CO-NR_{11}-, -O-CH_2-CO-NR_{12}-, -O-CH_2-CH_2-CZ- or -CH=CH-NR_{13}-,
R_3	is C_{1-6}alkyl, C_{1-6}haloalkyl, C_{1-6}cyanoalkyl, C_{1-6}alkoxy, C_{3-6}cycloalkoxy, C_{3-6}alkenyloxy, C_{3-6}alkynyloxy or C_{1-6}alkoxy substituted with cyano, halogen, C_{1-6}alkoxy, C_{1-6}alkoxycarbonyl, C_{3-6}alkynyloxycarbonyl or C_{1-4}alkoxy-C_{1-4}alkoxycarbonyl;
R_4	is hydrogen or C_{1-4}alkyl;
R_5	is C_{1-4}alkyl, C_{3-4}alkenyl, C_{3-4}alkynyl or C_{1-4}alkyl substituted with cyano, formyl or C_{1-4}alkoxycarbonyl;
R_6	is hydrogen or C_{1-4}alkyl;
R_7	is C_{1-4}alkylsulfonyl, C_{1-4}alkyl or C_{1-4}alkyl substituted by C_{1-4}alkoxycarbonyl;
R_8	is C_{1-4}alkyl;
R_9	is C_{1-4}alkyl, C_{1-4}cyanoalkyl, C_{1-4}alkoxy or C_{3-4}alkenyl; or
R_8 and R_9	together form a C_{4-5}alkylene bridge or an ethylene-O-ethylene bridge, which bridge may be substituted with C_{1-4}alkyl or C_{1-4}alkoxycarbonyl;
R_{10}	is hydrogen or C_{1-4}alkyl;
R_{11}	is hydrogen, C_{1-4}alkyl, C_{3-4}alkenyl, C_{3-4}alkynyl, or C_{3-4}alkenyl substituted with halogen or C_{1-4}alkoxycarbonyl;
R_{12}	is C_{1-4}alkoxy;

R_{13} is C_{1-4}alkyl, C_{3-4}alkenyl or C_{3-4}alkynyl,
Y is oxygen or sulfur, and
Z is oxygen or =N-C_{1-4}alkoxy,

are effective compounds for controlling undesired weeds.

The compounds of formula I have pronounced herbicidal activity against monocotyledonous and dicotyledonous weeds. A particularly high level of activity is observed against dicotyledonous weeds. Further, the compounds of formula I are well tolerated by most culture crop plants such as soybeans, sugarbeets, sunflower, and especially monocotyledonous crops such as sorghum, rice, maize and cereals, e.g. wheat barley, rye and oats.

In the above definitions under formula I the generic expressions designate the following more detailed chemical moieties.

Any alkyl group in the compound of formula I may be branched or straight chain and preferably has one to six carbon atoms, preferably one to four carbon atoms. Typical examples are methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, see-butyl or tert. butyl.

Any alkenyl or alkynyl group may be either branched or straight chain and preferably has two to six carbon atoms. Examples are vinyl, allyl, methallyl, 2-butenyl, ethinyl, propargyl, 2-butinyl, 2-pentenyl, 3-pentenyl, 3-butinyl, or 2-pentinyl.

Halo or halogen as used herein, refers to chlorine, fluorine, bromine and iodine, with fluorine and chlorine being preferred.

Any cycloalkyl group preferably has three to six carbon ring atoms. Preferred examples are cyclopropyl, cyclopentyl or cyclohexyl.

The alkyl portion of alkoxy, haloalkyl or cyanoalkyl have the same designation as given for alkyl above. In particular, examples are methoxy, ethoxy, isopropoxy, propoxy, butyloxy, cyanomethyl, 2-cyanoethyl, 1-cyanoethyl, 1-cyano-1-methylethyl, 3-cyanopropyl, chloromethyl, 2,2,2-trichloroethyl, trifluoromethyl, fluoromethyl, difluoromethyl, 1,1,2,2-tetrafluoroethyl, 2,2,2-trifluoroethyl, 2-chloroethyl or 2-fluoroethyl.

The carbons atom in 7-position and 8a-position of the perhydroimidazo[1,5a]pyridine ring system are asymmetrically substituted. The resulting products may therefore exist in diastereomeric forms. In most cases the product obtained from the given method of preparation will be in racemic form. The isomers may be separated by routine separation methods as known in the art. Additionally stereoselective routes of synthesis may provide predominantly the R- or S-form, depending on the employed starting materials. According to the present invention where not specially mentioned the compounds refer the mixtures as obtained from synthesis containing all isomers in varying ratios.

Among the compounds of formula I those are preferred wherein either

a) X is oxygen, or
b) R is fluorine, or
c) R_1 is chlorine, or
d) R_1 and R_2 together form the bridge -O-CH_2-$CONR_{11}$- or
e) R_2 is halogen, C_{1-6}alkoxycarbonyl-C_{1-4}alkyl or C_{1-6}alkoxycarbonyl-C_{2-6}alkenyl.

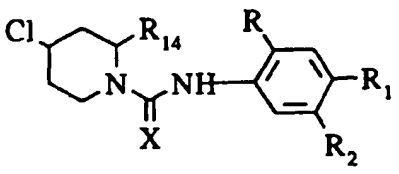
In an especially preferred subgroup of formula I X is oxygen, R is fluorine, R_1 is chlorine and R_2 is halogen, C_{1-6}alkoxycarbonyl-C_{1-4}alkyl or C_{1-6}alkoxycarbonyl-C_{2-4}alkenyl.

Preferred individual compounds of formula I are:

7-chloro-2-(5-bromo-4-chloro-2-fluorophenyl)-perhydroimidazo[1,5a]pyridine-1,3-dione;
methyl β-[2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-phenyl]-propionate; and
methyl 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-cinnamate.

Compounds of the formula I are useful because they demonstrate herbicidal activity, combined with excellent selectivity in cultivated crop plants.

Compounds of the formula I may be obtained from an intramolecular condensation of a compound of formula II



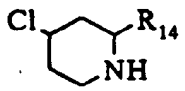
(II)

wherein

X, n, R, R_1 and R_2	are as defined for formula I and
R_{14}	is COOH, COOW or COSW, and
W	is C_{1-4}alkyl.

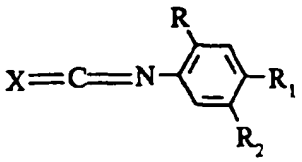
This condensation reaction is carried out under conditions that are typical for preparing hydantoin compounds. The reaction is facilitated by the presence of an acid or an alkaline agent. The condensation reaction may be carried out under acidic or alkaline conditions. Accordingly, the reaction may be carried out in an inert medium such as toluene in the presence of an alkaline agent such as triethylamine. Suitable temperatures range from about room to 60°C, preferably about 50°C. The resulting product is isolated and purified in accordance with known processes such as extraction and crystallization.

Compounds of the formula II may be prepared by reacting a compound of the formula III



(III)

wherein R_{14} is defined under formula II with a substituted phenyl isocyanate or isothiocyanate of formula IV



(IV)

wherein R, R_1, R_2 and X are as defined under formula II.

This reaction may be carried out in an inert medium such as toluene, preferably at ambient temperature. The resulting compound of formula II can be recovered from solution by standard separation techniques, e.g. suction filtration and chromatography.

The substituted phenyl isocyanates or isothiocyanates of formula IV are known. Compounds of the formula III are either known or can be prepared from known compounds according to known procedure. (c.f. Tetrahedron, Vol. 47 (24), 40394062, 1991).

The compounds of formula I are effective in controlling the growth of plants. By plants it is meant germinating seeds, emerging seedlings and established vegetation including underground portions. In particular, the compounds are useful as herbicides as indicated by causing damage to both monocotyledonous and dicotyledonous plants in various standard evaluations for determining such effects. The herbicidal effects are exhibited both pre- and post-emergence the plants. Such herbicidal effects indicate that the compounds of formula I are particularly of interest in combatting and controlling weeds (unwanted plants).

The compounds of the formula I are indicated mainly to be stronger acting against dicotyledonous plants than monocotyledonous plants. Relatively less toxicity towards crops than towards weeds is further indicated. Hence, the compounds are of particular interest as selective herbicides to combat weeds in a crop locus, particularly as locus of a crop such as, for example, sugarbeet, sunflower, cotton, soybean, rice, maize and cereals, e.g. wheat, barley, rye and oats, but especially wheat.

The present invention therefore also provides a method of combatting weeds in a locus which comprises applying to the weeds or their locus a herbicidally effective amount of a compound of the invention. When selective action is desired in crop locus, the amount applied will be sufficient to combat weeds without substantially damaging the crop.

For general herbicidal as well as selective herbicidal use of the compounds of the invention, the particular amounts to be applied will vary depending upon recognized factors such as the compound employed, the plants primarily in the locus, the timing, mode and formulation in application, the various conditions of treatment such as soil and weather and the like. However, in general, satisfactory results in weed control are usually obtained upon application of the compounds of the invention at a rate in the range of from 0.001 to 2 kg/hectare, more usually 0.01 to 1 kg/hectare, and preferably 0.01 to 0.25 kg/hectare, the application being repeated as necessary. When used in crops, the application usually will not exceed about 1 kg/hectare, and is usually in the range of 0.01 to 0.5 kg/hectare.

For practical use as herbicides, the compounds of formula I may be and are preferably employed in herbicidal compositions comprising a herbicidal effective amount of the compound and an inert carrier which is agriculturally acceptable in the sense of not, by reason of its presence, poisoning the agricultural environment including the immediate soil of application or any crops present therein or otherwise being unsafe for application. Such compositions of formulations may contain 0.01% to 99% by weight of active ingredient, from 0 to 20% by weight of agriculturally acceptable surfactants and 1 to 99.99% by weight of the inert carrier. Higher ratios of surfactant to active ingredient are sometimes desirable and are achieved by incorporation into the formulation or by tank mixing. Application forms of composition typically contain between 0.01 and 25% by weight of active ingredient, but lower or higher levels of active ingredient can, of course, be present depending on the intended use and the physical properties of the compounds. Concentrate forms of composition intended to be diluted before use generally contain between 2 and 90%, preferably between 10 and 80% by weight of active ingredient.

Useful compositions or formulations of the compounds of the invention include dusts; granules, pellets, suspension concentrates, wettable powders, emulsifiable concentrates and the like. They are obtained by conventional manner, e.g. by mixing the compounds of the invention with the inert carrier. More specifically, liquid compositions are obtained by mixing the ingredients, fine solid compositions by blending and, usually grinding, suspensions by wet milling and granules and pellets by impregnating or coating (preformed) granular carriers with the active ingredient or by agglomeration techniques.

For example, dusts can be prepared by grinding and blending the active compound with a solid inert carrier such as talc, clay, silica and the like. Granular formulations can be prepared by impregnating the compound, usually dissolved in a suitable solvent, onto and into granulated carriers such as the attapulgites or the vermiculites, usually of a particle size range of from about 0.3 to 1.5 mm. Wettable powders, which can be dispersed in water or oil to any desired concentration of the active compound, can be prepared by incorporating wetting agents into concentrated dust compositions.

Alternatively, the compounds of the invention may be used in micro-encapsulated form.

Agriculturally acceptable additives may be employed in the herbicidal compositions to improve the performance of the active ingredient and to reduce foaming, caking and corrosion.

Surfactant as used herein means agriculturally acceptable material which imparts emulsifiability, spreading, wetting, disperibility or other surface-modifying properties. Examples of surfactants are sodium lignin sulphonate and lauryl sulphate.

Carriers as used herein mean a liquid or solid material used to dilute a concentrated material to a usable or desirable strength. For dusts or granules it can be e.g. talc, kaolin or diatomaeous earth, for liquid concentrate forms, a hydrocarbon such as xylene or an alcohol such as isopropanol; and for liquid application forms, e.g. water or diesel oil.

The compositions of this application can also comprise other compounds having biological activity, e.g. compounds having similar or complementary herbicidal activity or compounds having antidotal, fungicidal or insecticidal activity.

Typical herbicidal composition, according to this invention, are illustrated by the following Examples A, B and C in which the quantities are in parts by weight.

EXAMPLE A

Preparation of a Dust

10 Parts of a compound of formula I and 90 parts of powdered talc are mixed in a mechanical grinder-blender and are ground until a homogenous, free flowing dust of the desired particle size is obtained. This dust is suitable for direct application to the site of the weed infestation.

EXAMPLE B

Preparation of Wettable Powder

25 Parts of a compound of formula I are mixed and milled with 25 parts of synthetic fine silica, 2 parts of sodium lauryl sulphate, 3 parts of sodium lignin sulphonate and 45 parts of finely divided kaolin until the mean particle size is about 5 micron. The resulting wettable powder is diluted with water before use to a spray liquor with the desired concentration.

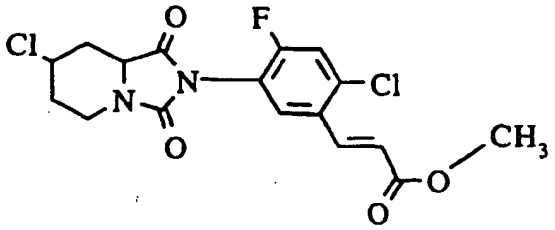
EXAMPLE C

Preparation of Emulsifiable Concentrate (EC)

13 Parts of a compound of formula I are mixed in a beaker with 7 parts of Toximul 360A (a mixture of anionic and non-ionic surfactants containing largely non-ionic surfactants), 24 parts of dimethylformamide and 56 parts of Tenneco 500-100 (predominantly a mixture of alkylated aromatics such as xylene and ethylbenzene) until solution is effected. The resulting EC is diluted with water for use.

EXAMPLE 1

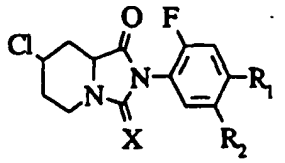
Methyl 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)cinnamate



To a solution of 38,5g (0,18 mol)4-chloro-2-piperidine carboxylic acid methyl ester hydrochloride and 40g (0,396 mol) triethylamine in 1200 ml CH_2Cl_2 is added in small portions 46g (0,18 mol) isocyanate of methyl 2-chloro-4-fluoro-5-amino-cinnamate dissolved in 300 ml CH_2Cl_2. The reaction solution is stirred at room temperature for a period of 20 hours, washed with 2 x 500 ml water, dried with Na_2SO_4 and the solvent is evaporated. The residual brown syrup is treated with 500 ml ether/hexane (4:1) affording methyl 2-chloro4-fluoro-5-(7-chloro-perhydroimidazo[1,5]pyridine-1,3-dione-2-yl) cinnamate as a white powder with m.p. of 162-163°C.

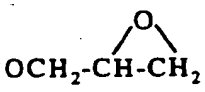
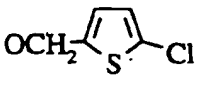
The compounds of the following tables are obtained in analogous manner.

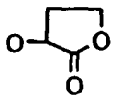
Table 1:

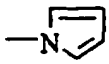
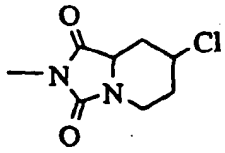
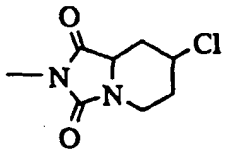


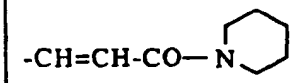
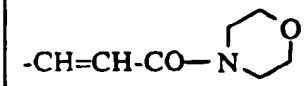
No.	R_1	R_2	X	mp. or Rf on SiO_2
1.01	CH_3	$COOCH(CH_3)_2$	O	130-131°C
1.02	Cl	$COOCH_3$	O	0,12 HXF/EST 7:3
1.03	Cl	$COOCH_2CH_3$	O	82-85°C
1.04	Cl	$COOCH_2CN$	O	176-178°C
1.05	Cl	$COOCH_2CH_2CH_3$	O	0.21 HXF/EST 7:3
1.06	Cl	$COOCH_2CH_2CH_2CH_3$	O	0.23 HXF/EST 7:3
1.07	Cl	$COOCH(CH_3)CH_2CH_3$	O	0.26 HXF/EST 7:3
1.08	Cl	$COOC(CH_3)_3$	O	157°C
1.09	Cl	$COOCH_2CH(CH_3)_2$	O	0.26 HXF/EST 7:3
1.10	Cl	$COOCH_2CH_2OCH_2CH_3$	O	0.28 HXF/EST 1:1
1.11	Cl	$COOCH(CH_3)CH_2OCH_3$	O	0.11 HXF/EST 7:3
1.12	Cl	$COO\text{-}C_5H_9$-cycl.	O	0.23 HXF/EST 7:3
1.13	Cl	$COO(CH_2)_4CH_3$	O	0.27 HXF/EST 7:3
1.14	Cl	$COOC(CH_3)_2CH=CH_2$	O	0.24 HXF/EST 7:3
1.15	Cl	$COOCH_2CH_2OCH_3$	O	0.27 HXF/EST 1:1
1.16	Cl	$COOCH(CH_3)$-CN	O	0.10 HXF/EST 7:3
1.17	Cl	$COOCH_2CH=CH_2$	O	123-125°C

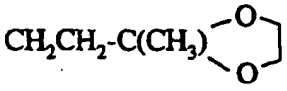
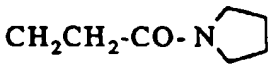
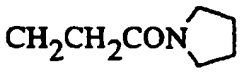
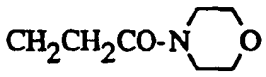
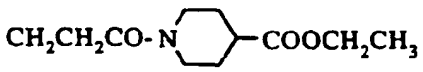
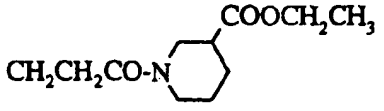
1.18	Cl	$COOCH_2C{\equiv}CH$	O	128-130°C
1.19	Cl	$COOCH_2$-$C(CH_3){=}CH_2$	O	0.15 SE/EST 1:1
1.20	Cl	$COOCH_2CH{=}CH$-CH_3	O	0.17 SE/EST 1:1
1.21	Cl	$COOCH(CH_3)_2$	O	0.22 HXF/EST 7:3
1.22	Cl	$COOCH(CH_3)$-$CH(CH_3)_2$	O	0.35 HXF/EST 7:3
1.23	Cl	$COOCH(CH_3)CH_2CH_2CH_3$	O	0.36 HXF/EST 7:3
1.24	Cl	$COOCH(CH_3)CH_2CH{=}CH_2$	O	0.23 HXF/EST 7:3
1.25	Cl	$COOCH(CH_3)CH_2COOCH_3$	O	0.10 HXF/EST 7:3
1.26	Cl	$COOCH(CH_3)CH_2CH(CH_3)_2$	O	0.27 HXF/EST 7:3
1.27	Cl	$COOCH(CH_3)CH_2CH_2CH_2CH_3$.	O	0.28 HXF/EST 7:3
1.28	Cl	$COOC(CH_3)_2$-CN	O	58°C
1.29	Cl	COOH	O	196-199°C
1.30	Cl	$COOCH_3$	S	149-151°C
1.31	Cl	$COOCH_2CH_3$	S	161-162°C
1.32	Cl	$COOCH(CH_3)_2$	S	163-165°C
1.33	Cl	$COOCH_2CH_2OCH_3$	S	138-140°C
1.34	Cl	$COOCH(CH_3)CH_2OCH_3$	S	111-113°C
1.35	Cl	$CON(CH_3)$-$CH(CH_3)_2$	O	166-168°C
1.36	Cl	$CON(CH_3)_2$	O	185-187°C
1.37	Cl	$CONH(CH_2)_3CH_3$	O	0.15 HXF/EST 1:1
1.38	Cl	$CONHC(CH_3)_3$	O	0.30 HXF/EST 1:1
1.39	Cl	$CON(CH_3)$-$(CH_2)_3CH_3$	O	0.10 HXF/EST 1:1
1.40	Cl	$CON(CH_3)CH_2CH_2C{\equiv}N$	O	0.08 HXF/EST 3:7

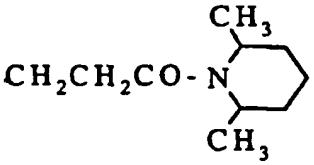
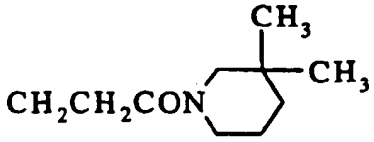
1.41	Cl	$CON(CH_3)CH_2C{\equiv}CH$	O	0.18 HXF/EST 1:1
1.42	Cl	$CON(CH_2CH_3)_2$	O	0.22 HXF/EST 3:7
1.43	Cl	$CON(CH_3)CH_2COOCH_3$	O	0.20 HXF/EST 3:7
1.44	Cl	$CON(CH_3)CH_2CH{=}O$	O	0.07 HXF/EST 3:7
1.45	Br	OCH_3	O	116-119°C
1.46	Br	OCH_2CH_3	O	209-211°C
1.47	Br	$OCH_2CH{=}CH\text{-}Cl$	O	205-208°C
1.48	Br	$O(CH_2)_3CN$	O	59-62°C
1.49	Cl	O-benzyl	O	256-258°C
1.50	Cl	$OCH(CH_3)_2$	S	134°C
1.51	Cl	$OCH(CH_3)_2$	O	96-98°C
1.52	Cl	$OCH_2CH_2OCH_3$	O	111-113°C
1.53	Cl	$OCH_2CH_2OCH_2CH_3$	O	0.17 HXF/EST 1:1
1.54	Cl	$OCH(CH_3)\text{-}C{\equiv}CH$	O	142-144°C
1.55	Cl	$OCH_2C{\equiv}CH$	O	179-181°C
1.56	Cl	$OCH_2CH{=}CH_2$	O	0.17 HXF/EST 7:3
1.57	Cl	$OCH_2\text{-}C_3H_5\text{-cycl.}$	O	59-60°C
1.58	Cl		O	63°C
1.59	Cl		O	173-175°C
1.60	Cl	$OCH_2CH{=}CH\text{-}C{\equiv}C\text{-}C(CH_3)_3$	O	0.24 HXF/EST 7:3

1.61	Cl	-O-C_5H_9-cycl.	O	0.38 HXF/EST 2:8
1.62	CN	OCH_2CH_3	O	290-292°C
1.63	CN	F	O	265-268°C
1.64	CH_3	$OCH_2COO(CH_2)_4CH_3$	O	82°C
1.65	Cl	OCH_2COOCH_3	O	0.24 HXF/EST 1:1
1.66	Cl	$OCH_2COOCH_2CH_3$	O	0.22 SE/HXF 8:2
1.67	Cl	$OCH_2COOCH_2CH_2CH_3$	O	0.20 SE/HXF 7:3
1.68	Cl	$OCH_2COOCH(CH_3)_2$	O	0.16 SE/HXF 7:3
1.69	Cl	$OCH_2COO(CH_2)_3CH_3$	O	0.21 SE/HXF 7:3
1.70	Cl	$OCH_2COO(CH_2)_4CH_3$	O	0.45 HXF/EST 1:1
1.71	Cl	$OCH(CH_3)COOCH_3$	O	0.20 SE/HXF 8:2
1.72	Cl	$OCH(CH_3)COOCH_2CH_3$	O	0.13 HXF/EST 7:3
1.73	Cl	$OCH(CH_3)COOCH_2CH_2CH_3$	O	0.15 HXF/EST 7:3
1.74	Cl	$OCH(CH_3)COOCH(CH_3)_2$	O	0.27 SE/HXF 7:3
1.75	Cl	 Isomer 1 Isomer 2	O O	0.38 EST 0.43 EST
1.76	Cl	$OCOC(CH_3)_3$	O	0.16 SE/HXF 1:1
1.77	Cl	$OCH_2OCOC(CH_3)_3$	O	0.17 SE/HXF 1:1
1.78	Cl	$CH(CH_3)_2$	O	146-148°C
1.79	Cl	$CH_2OCH_2COOCH_3$	O	0.06 HXF +2.5% EST
1.80	Cl	CH_2OCOCH_3	O	0.37 HXF/EST 1:1
1.81	Cl	$CH_2OCOCH_2CH_3$	O	0.07 HXF +20% EST

1.82	Cl	$CH_2OCOCH(CH_3)_2$	O	0.10 HXF +30% EST
1.83	Cl	CH_2OCOCH_2Cl	O	0.08 HXF +30% EST
1.84	CH_3	$CH(CH_3)OCOCH_3$	O	0.15 HXF +30% EST
1.85	CH_3	CH_2CH_3	O	0.1 HXF +2.5% EST
1.86	Cl	Br	O	177-179°C
1.87	Cl	J	O	183-184°C
1.88	Br	OCH_2-$C(CH_3)_3$	O	282-284°C
1.89	Cl	NH-CH_3	O	0.17 HXF/EST 6:4
1.90	Cl	$N(CH_3)2$	O	0.26 SE
1.91	Cl		O	0.15 SE/HXF 1:1
1.92	Cl		O	0.11 EST/HXF 1:1
1.93	F		O	0.13 EST/HXF 1:1
1.94	Cl	-CH=CH-COOH	O	239-242°C
1.95	Cl	-CH=CH-$COOCH_3$	O	162-163°C
1.96	Cl	-CH=CH-$COOCH_2CH_3$	O	76-78°C
1.97	Cl	-CH=CH-$COOCH_2CH_2CH_3$	O	75-77°C
1.98	Cl	-CH=CH-$COOCH(CH_3)_2$	O	78-80°C

1.99	Cl	-CH=CH-COOCH(CH_3)CH_2CH_3	O	121-122°C
1.100	Cl	-CH=CH-COO(CH_2)$_4CH_3$	O	86-88°C
1.101	Cl	-CH=CH-COOCH-CH_3 | COOCH-CH_3 | CH_2-OCH_3	O	88-89°C
1.102	Cl	-CH=CH-COOCH(CH_3)$COOCH_2C\equiv CH$	O	123-125°C
1.103	Cl	-CH=CH-$COOCH_2COOCH_2CH(CH_3)_2$	O	72-74°C
1.104	Cl	-CH=CH-$COCH_3$	O	152-153°C
1.105	Cl	-CH=CH-CH(CH_3)-OCH_3	O	112-114°C
1.106	Cl	-CH=CH-CON(CH_3)-$CH(CH_3)_2$	O	202-204°C
1.108	Cl	-CH=CH-$CON[CH(CH_3)_2]_2$	O	167-168°C
1.109	Cl	-CH=CH-CON(CH_3)$CH_2CH_2C\equiv N$	O	196-197°C
1.110	Cl		O	128-130°C
1.111	Cl		O	136-138°C
1.112	Cl		O	237-238°C
1.113	Cl	-CH=CH-CON(CH_3)OCH_3	O	170-172°C
1.114	Cl	-CH_2-CH_2COOH	O	174-172°C
1.115	Cl	CH_2CH_2-$COOCH_3$	O	110-112°C

1.116	Cl	CH_2-CHCl-$COOCH_3$	O	0.33 EST/HXF 1:1
1.117	Cl	CH_2CH_2-$COOCH_2CH_3$	O	0.40 EST/HXF 1:1
1.118	Cl	CH_2CH_2-$COOCH_2CH_2CH_3$	O	0.36 EST/HXF 1:1
1.119	Cl	CH_2CH_2-$COOCH(CH_3)_2$	O	0.48 EST/HXF 2:1
1.120	Cl	CH_2CH_2-$COOCH(CH_3)CH_2CH_3$	O	0.38 EST/HXF 1:1
1.121	Cl	$CH_2CH_2COCH_3$	O	163-165°C
1.122	Cl	$CH_2CH_2C(CH_3)$=NOH	O	95-96°C
1.123	Cl	$CH_2CH_2C(CH_3)$=$NOCH_3$	O	104-105°C
1.124	Cl		O	149-150°C
1.125	Cl		O	0.11 EST
1.126	Cl		O	0.26 EST
1.127	Cl		O	0.15 EST
1.128	Cl	CH_2CH_2-$CON(CH_3)CH_2CH_2CN$	O	0.20 EST
1.129	Cl	CH_2CH_2-$CON(CH_3)CH_2CH$=CH_2	O	0.28 EST
1.130	Cl		O	0.32 EST
1.131	Cl		O	0.31 EST

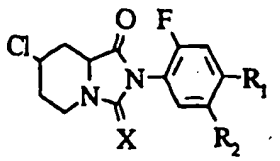
1.132	Cl		O	0.39 EST
1.133	Cl		O	0.35 EST
1.134	Cl	$CH_2CH(CH_3)CH_2Cl$	O	88-90°C
1.135	Cl	-CH=CCl-$COOCH_3$	O	
1.136	Cl	-CH=CCl-$COOC_2H_5$	O	
1.137	Cl	NH-CH_2-$COOCH_3$	O	
1.138	Cl	NH-$CH(CH_3)$-$COOCH_3$	O	
1.139	Cl	N(CH_3)-CH_2-$COOCH_3$	O	
1.140	Cl	N(CH_3)-$CH(CH_3)$-$COOCH_3$	O	
1.141	Cl	N(CH_3)-SO_2-CH_3	O	
1.142	Cl	N(CH_3)-SO_2-C_2H_5	O	
1.143	Br	O-CH_2-$C(CH_3)$=CH_2	O	0.2 EST/ HXF 3:7

HXF = hexane

EST = ethyl acetate

SE = diethyl ether

Table 2:



No.	$R_1 + R_2$	X	m.p. or Rf on SiO_2
2.01	-O-CH_2-CO-NH-	S	223-224°C
2.02	-O-CH_2-CO-N- | $CH_2C{\equiv}CH$	O	198-200°C
2.03	-O-CH_2-CO-N-$CH_2CH{=}CH_2$ |	O	208-109°C
2.04	-O-CH_2-CO-N-$CH_2C{\equiv}CH$ |	S	146-148°C
2.05	-OCH(CH_3)-CO-N-$CH_2C{\equiv}CH$ |	O	130-134°C
2.06	-S-CH_2-CO-N-$CH_2C{\equiv}CH$ |	O	243-244°C
2.07	CH_3 | -O-CH_2-CO-N-CH_2-C=CH_2 |	O	165-167°C
2.08	Br | -O-CH_2-CO-N-CH_2-C=CH_2 |	O	210-211°C

2.09	-O-CH_2-CO-N-CH_2CH=CH-$COOCH_3$ |	O	240-242°C
2.10	-O-CH_2-CO-N-CH_2CH=CH-Br |	O	206-208°C
2.11	-O-CH_2-CO-N-CH_2CH=CH-CH_3 |	O	163-165°C
2.12	Cl | -O-CH_2-CO-N-CH_2-C=CH_2 |	O	208-210°C
2.13	-O-CH_2CO-N-CH_2CH_3 |	O	214-216°C
2.14	-O-CH_2-CO-N-CH_3 |	O	239-240°C
2.15	-O-CH_2-CH_2-CO-	O	213-214°C
2.16	-O-CH_2-CH_2-C=NOCH_3 |	O	169°C
2.17	-O-CH_2-CO-N-OCH_2CH_3 |	O	221°C
2.18	-CH=CH-N-CH_2C≡CH |	O	172-173°C

Biological Test

Example H1: Post-emergence Herbicidal Activity

Pots filled with a sandy loam type of soil are sown with 14 dicotyledonous weeds and kept under standard germination conditions in a greenhouse.

When the plants have about 2 leaves they are sprayed at 3 or 4 rates (5.5, 16.6, 50 and 150 g ai/ha) with a spray volume of 600 l/ha. The pots are watered as needed from above. The herbicidal activity is visually estimated (0-100%, in comparison to an untreated control) 25 days after application.

In this test the compounds of formula I exhibited good herbicidal activity. Compounds 1.17, 1.22, 1.25, 1.31, 1.38,

1.61, 1.63, 1.68, 1.72, 1.86, 1.110, 1.113, 1.117, 1.126, 1.143 and 2.14 showed more than 90% efficacy at an application rate of 50 g a.i./ha under the test conditions.

Example H2: Comparative Test

In a postemergence test carried out in a green house pots were filled with a sandy loam type of soil are sown with Abutilon theophrasti, Galium aparine, Ipomoea purpurea, Matricaria chamomilla, Portulaca oleracea, Senecio vulgaris, Solanum nigrum, Stellaria media, Xanthium strumarium, maize and wheat. When the plants have about 2 leaves they are sprayed at rates of 16.6 g ai/ha with a spray volume of 600 l/ha. The pots are watered as needed from above. The herbicidal activity is visually estimated (0-100%, in comparison to an untreated control) 25 days after application.

In order to compare efficacy and selectivity of the compounds of present invention to the compounds of EP-A-493 323 representative species were tested in the above test procedure.

The tested compounds were:

Compounds 1.08, 1.21 and 1.51 from present application and compounds A (known from EP-A-493323 as compound 1.53), B (known from EP-A493323 as compound 1.34), and C (known from EP-A-493323).

The chemical designations of the test compounds are:

Compound 1.08	tert. butyl 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl) benzoate,
Compound A	tert.butyl 2-chloro-4-fluoro-5-(7-fluoro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)benzoate,
Compound 1.21	isopropyl 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl) benzoate,
Compound B	isopropyl 2-chloro-4-fluoro-5-(7-fluoro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)benzoate,
Compound 1.51	7-chloro-2-(4-chloro-2-fluoro-5-isopropoxyphenyl)-perhydroimidazo[1,5a]pyridine-1,3-dione,
Compound C	7-fluoro-2-(4-chloro-2-fluoro-5-isopropoxyphenyl)-perhydroimidazo[1,5a]pyridine-1,3-dione.

The following test results were obtained (plant damage in %):

a) post-emergence: maize, Variety Mutin
application rate 16.6 g a.i./ha.

plant	Compound 1.08	Compound A
maize	0	20
abutilon	100	100
chenopodium	100	100
ipomoea	100	90
portulaca	100	100
solanum	100	100
xanthium	100	100

plant	Compound 1.21	Compound B
maize	20	40
abutilon	100	100
chenopodium	100	100
ipomoea	100	100
portulaca	100	100
solanum	100	100
xanthium	100	100

plant	Compound 1.51	Compound C
maize	20	70
abutilon	100	100
chenopodium	100	100
ipomoea	100	100
portulaca	100	100
solanum	100	100
xanthium	100	100

b) post-emergence: wheat, Variety Albis
application rate 16.6 g a.i/ha

plant	Compound 1.08	Compound A
wheat	80	100
chenopodium	100	100
galium	100	80
matricaria	100	-
senecio	95	80
stellaria	100	100

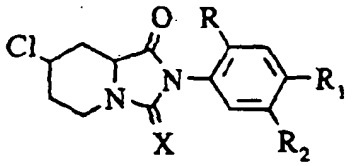
plant	Compound 1.21	Compound B
wheat	30	40
chenopodium	100	100
galium	100	100
matricaria	100	-
senecio	100	100
stellaria	100	100

plant	Compound 1.51	Compound C
wheat	20	70
chenopodium	100	100
galium	100	100
matricaria	100	90
senecio	80	90
stellaria	100	40

From all comparative tests it is evident that the chloro-compounds of present invention are better tolerated by maize and wheat plants than the fluoro-compounds known from the prior art. For practical applications in wheat compound 1.08 will require application rates lower than the tested rate.

Claims

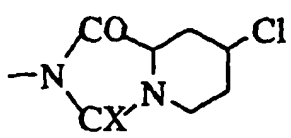
1. A compound of formula I



(I)

wherein

X is oxygen or sulfur,
R is hydrogen, chlorine or fluorine,
R_1 is fluorine, chlorine, bromine, cyano or methyl,
R_2 is halogen, C_{1-6}alkyl, C_{1-6}alkoxy, C_{1-6}alkylcarbonyloxy, C_{3-6}cycloalkoxy, C_{3-6}alkynyloxy, C_{3-6}alkenyloxy, -COOH, -CO-R_3, -CO-NR_4R_5, -NR_6R_7, C_{2-6}alkenyl, N-pyrrolyl, 2-oxo-3-tetrahydrofuranyl or a group



, or is
C_{1-6}alkyl substituted with halogen, COOH, -CO-R_3, -CO-NR_8R_9, =N-OH, =N-C_{1-4}alkoxy, -O-CO-R_3 or -O-C_{2-3}alkylene-O-; or is
C_{2-6}alkenyl substituted with halogen, COOH, -CO-R_3, -CO-NR_8R_9 or C_{1-4}alkoxy; or is C_{1-6}alkoxy substituted with cyano, -CO-R_3, C_{1-4}alkoxy, C_{3-6}cycloalkyl, oxiranyl, or optionally substituted thienyl

	or phenyl with the substituents selected from C_{1-6}alkyl, C_{1-4}alkoxy or halogen; or is C_{3-6}alkenyloxy substituted by C_{2-5}alkynyl; or
R_1 and R_2	together form a bridge member selected from -Y-CHR_{10}-CO-NR_{11}-, -O-CH_2-CO-NR_{12}-, -O-CH_2-CH_2-CZ- or -CH=CH-NR_{13}-,
R_3	is C_{1-6}alkyl, C_{1-6}haloalkyl, C_{1-6}cyanoalkyl, C_{1-6}alkoxy, C_{3-6}cycloalkoxy, C_{3-6}alkenyloxy, C_{3-6}alkynyloxy or C_{1-6}alkoxy substituted with cyano, halogen, C_{1-6}alkoxy, C_{1-6}alkoxycarbonyl, C_{3-6}alkynyloxycarbonyl or C_{1-4}alkoxy-C_{1-4}alkoxycarbonyl;
R_4	is hydrogen or C_{1-4}alkyl;
R_5	is C_{1-4}alkyl, C_{3-4}alkenyl, C_{3-4}alkinyl or C_{1-4}alkyl substituted with cyano, formyl or C_{1-4}alkoxycarbonyl;
R_6	is hydrogen or C_{1-4}alkyl;
R_7	is C_{1-4}alkylsulfonyl, C_{1-4}alkyl or C_{1-4}alkyl substituted by C_{1-4}alkoxycarbonyl;
R_8	is C_{1-4}alkyl;
R_9	is C_{1-4}alkyl, C_{1-4}cyanoalkyl, C_{1-4}alkoxy or C_{3-4}alkenyl; or
R_8 and R_9	together form a C_{4-5}alkylene bridge or a ethylene-O-ethylene bridge, which bridge may be substituted with C_{1-4}alkyl or C_{1-4}alkoxycarbonyl;
R_{10}	is hydrogen or C_{1-4}alkyl;
R_{11}	is hydrogen, C_{1-4}alkyl, C_{3-4}alkenyl, C_{3-4}alkynyl, or C_{3-4}alkenyl substituted with halogen or C_{1-4}alkoxycarbonyl;
R_{12}	is C_{1-4}alkoxy;
R_{13}	is C_{1-4}alkyl, C_{3-4}alkenyl or C_{3-4}alkynyl,
Y	is oxygen or sulfur, and
Z	is oxygen or =N-C_{1-4}alkoxy.

2. A compound according to claim 1 wherein X is oxygen.

3. A compound according to any one of claims 1 or 2 wherein R is fluorine and R_1 is chlorine.

4. A compound according to claim 1, wherein R_1 and R_2 together form the bridge -O-CH_2-CO-NR_{11}-.

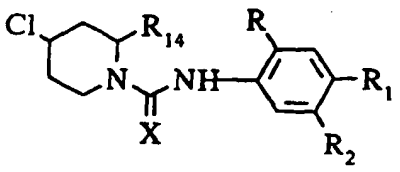
5. A compound according to claim 1, wherein R_2 is halogen, C_{1-6}alkoxycarbonyl-C_{1-4}alkyl or C_{1-6}alkoxycarbonyl-C_{2-6}alkenyl.

6. A compound according to claim 1, wherein X is oxygen, R is fluorine, R_1 is chlorine and R_2 is halogen, C_{1-6}alkoxycarbonyl-C_{1-4}alkyl -C_{1-6}alkyl or C_{1-6}alkoxycarbonyl-C_{2-4}alkenyl.

7. A compound according to claim 1 selected from the group comprising

7-chloro-2-(5-bromo-4-chloro-2-fluorophenyl)-perhydroimidazo[1,5a]pyridine-1,3-dione;
methyl β-[2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-phenyl]-propionate; and
methyl 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-cinnamate.

8. A process for the preparation of a compound of formula I according to claim 1, which comprises intramolecular condensation of a compound of formula II



(II)

wherein X, n, R, R_1 and R_2 are as defined in Claim 1, R_{1-4} is COOH, COOW and COSW, and W is C_{1-4}alkyl.

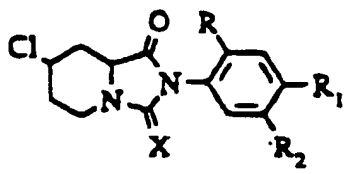
9. A herbicidal composition comprising a compound of formula I according to Claim 1 and an agriculturally acceptable

carrier.

10. A method of combatting weeds which comprises applying to the weeds or their locus a herbicidally effective amount of a compound of formula I according to Claim 1.

Patentansprüche

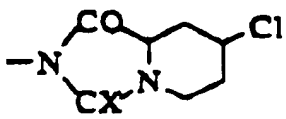
1. Verbindung der Formel I



(I)

worin

X Sauerstoff oder Schwefel darstellt,
R Wasserstoff, Chlor oder Fluor darstellt,
R_1 Fluor, Chlor, Brom, Cyano oder Methyl darstellt,
R_2 Halogen, C_{1-6}-Alkyl, C_{1-6}-Alkoxy, C_{1-6}-Alkylcarbonyloxy, C_{3-6}-Cycloalkoxy, C_{3-6}-Alkinyloxy, C_{3-6}-Alkenyloxy, -COOH, -CO-R_3, -CO-NR_4R_5, -NR_6R_7, C_{2-6}-Alkenyl, N-Pyrrolyl, 2-Oxo-3-tetrahydrofuranyl oder eine Gruppe



darstellt,
oder
C_{1-6}-Alkyl, substituiert mit Halogen, COOH, -CO-R_3, -CO-NR_8R_9, =N-OH, =N-C_{1-4}-Alkoxy, -O-CO-R_3 oder -O-C_{2-3}-Alkylen-O-; oder
C_{2-6}-Alkenyl, substituiert mit Halogen, COOH, -CO-R_3, -CO-NR_8R_9 oder C_{1-4}-Alkoxy; oder
C_{1-6}-Alkoxy, substituiert mit Cyano, -CO-R_3, C_{1-4}-Alkoxy, C_{3-6}-Cycloalkyl, Oxiranyl, oder einer gegebenenfalls substituierten Thienyl- oder Phenylgruppe, wobei die Substituenten ausgewählt sind aus C_{1-4}-Alkyl, C_{1-4}-Alkoxy oder Halogen; oder
C_{3-6}-Alkenyloxy, substituiert mit C_{2-5}-Alkinyl, darstellt; oder
R_1 und R_2 zusammen ein Brückenglied, ausgewählt aus -Y-CHR_{10}-CO-NR_{11}-, -O-CH_2-CO-NR_{12}-, -O-CH_2-CH_2-CZ- oder -CH=CH-NR_{13}-, bilden,
R_3 C_{1-6}-Alkyl, C_{1-6}-Halogenalkyl, C_{1-6}-Cyanoalkyl, C_{1-6}-Alkoxy, C_{3-6}-cycloalkoxy, C_{3-6}-Alkenyloxy, C_{3-6}-Alkinyloxy oder C_{1-6}-Alkoxy, substituiert mit Cyano, Halogen, C_{1-6}-Alkoxy, C_{1-6}-Alkoxycarbonyl, C_{3-6}-Alkinyloxycarbonyl oder C_{1-4}-Alkoxy-C_{1-4}-alkoxycarbonyl darstellt;
R_4 Wasserstoff oder C_{1-4}-Alkyl darstellt;
R_5 C_{1-4}-Alkyl, C_{3-4}-Alkenyl, C_{3-4}-Alkinyl oder C_{1-4}-Alkyl, substituiert mit Cyano, Formyl oder C_{1-4}-Alkoxycarbonyl, darstellt;
R_6 Wasserstoff oder C_{1-4}-Alkyl darstellt;
R_7 C_{1-4}-Alkylsulfonyl, C_{1-4}-Alkyl oder C_{1-4}-Alkyl, substituiert mit C_{1-4}-Alkoxycarbonyl, darstellt;
R_8 C_{1-4}-Alkyl darstellt;
R_9 C_{1-4}-Alkyl, C_{1-4}-Cyanoalkyl, C_{1-4}-Alkoxy oder C_{3-4}-Alkenyl darstellt; oder
R_8 und R_9 zusammen eine C_{4-5}-Alkylenbrücke oder eine Ethylen-O-ethylenbrücke bilden, wobei die Brücke mit C_{1-4}-Alkyl oder C_{1-4}-Alkoxycarbonyl substituiert sein kann;
R_{10} Wasserstoff oder C_{1-4}-Alkyl darstellt;
R_{11} Wasserstoff, C_{1-4}-Alkyl, C_{3-4}-Alkenyl, C_{3-4}-Alkinyl oder C_{3-4}-Alkenyl, substituiert mit Halogen oder C_{1-4}-

Alkoxycarbonyl, darstellt;
R_{12} C_{1-4}-Alkoxy darstellt;
R_{13} C_{1-4}-Alkyl, C_{3-4}-Alkenyl oder C_{3-4}-Alkinyl darstellt,
Y Sauerstoff oder Schwefel darstellt und
Z Sauerstoff oder =N-C_{1-4}-Alkoxy darstellt.

2. Verbindung nach Anspruch 1, worin X Sauerstoff darstellt.

3. Verbindung nach einem der Ansprüche 1 oder 2, worin R Fluor darstellt und R_1 Chlor darstellt.

4. Verbindung nach Anspruch 1, worin R_1 und R_2 zusammen die Brücke -O-CH_2-CO-NR_{11}- bilden.

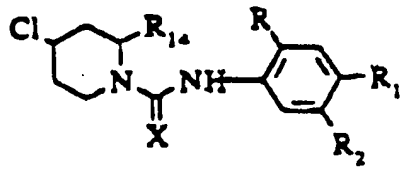
5. Verbindung nach Anspruch 1, worin R_2 Halogen, C_{1-6}-Alkoxycarbonyl-C_{1-4}-alkyl oder C_{1-6}-Alkoxycarbonyl-C_{2-6}-alkenyl darstellt.

6. Verbindung nach Anspruch 1, worin X Sauerstoff darstellt, R Fluor darstellt, R_1 Chlor darstellt und R_2 Halogen, C_{1-6}-Alkoxycarbonyl-C_{1-4}-alkyl oder C_{1-6}-Alkoxycarbonyl-C_{2-4}-alkenyl darstellt.

7. Verbindung nach Anspruch 1, ausgewählt aus der Gruppe, umfassend 7-Chlor-2-(5-brom-4-chlor-2-fluorphenyl)-perhydroimidazo[1,5a]pyridin-1,3-dion;

β-[2-Chlor-4-fluor-5-(7-chlorperhydroimidazo[1,5a]-pyridin-1,3-dion-2-yl)phenyl]propionsäuremethylester
und
2-Chlor-4-fluor-5-(7-chlorperhydroimidazo[1,5a]pyridin-1,3-dion-2-yl)-zimtsäuremethylester.

8. Verfahren zur Herstellung einer Verbindung der Formel I nach Anspruch 1, das intramolekulare Kondensation einer Verbindung der Formel II



(II)

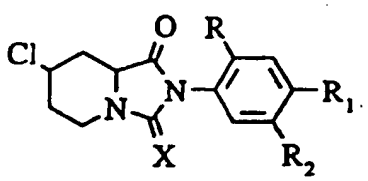
worin X, n, R, R_1 und R_2 wie in Anspruch 1 definiert sind, R_{14} COOH, COOW und COSW darstellt und W C_{1-4}-Alkyl darstellt, umfaßt.

9. Herbizides Mittel, umfassend eine Verbindung der Formel I nach Anspruch 1 und einen landwirtschaftlich verträglichen Träger.

10. Verfahren zum Bekämpfen von Unkräutern, das Applizieren einer herbizid wirksamen Menge einer Verbindung der Formel I nach Anspruch 1 auf die Unkräuter oder deren Ort umfaßt.

Revendications

1. Un composé de formule I



(I)

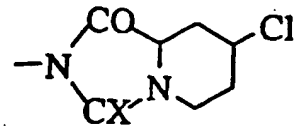
dans laquelle

X signifie l'oxygène ou le soufre,

R signifie l'hydrogène, le chlore ou le fluor,

R_1 signifie le fluor, le chlore, le brome, un groupe cyano ou méthyle,

R_2 signifie un halogène, un groupe C_{1-6}alkyle, C_{1-6}alcoxy, C_{1-6}alkylcarbonyloxy, C_{3-6}cycloalcoxy, C_{3-6}alcynyloxy, C_{3-6}alcényloxy, -COOH, -CO-R_3, -CO-NR_4-R_5, -NR_6R_7, C_{2-6}alcényle, N-pyrrolyle, 2-oxo-3-tétrahydrofuranyle ou un groupe



ou bien signifie
un groupe C_{1-6}alkyle substitué par un halogène, COOH, -CO-R_3, -CO-NR_8R_9, =N-OH, =N-C_{1-4}alcoxy, -O-CO-R_3 ou -O-C_{2-3}alkylène-O-; ou bien signifie
un groupe C_{2-6}alcényle substitué par un halogène, COOH, -CO-R_3, -CO-NR_8R_9 ou C_{1-4}alcoxy; ou bien signifie un groupe C_{1-6}alcoxy substitué par un groupe cyano, -CO-R_3, C_{1-4}alcoxy, C_{3-6}cycloalkyle, oxiranyle ou un groupe thiényle ou phényle éventuellement substitués, les substituants étant choisis parmi un groupe C_{1-4}alkyle, C_{1-4}alcoxy ou un halogène; ou bien signifie
C_{3-6}alcényloxy substitué par un groupe C_{2-5}alcynyle; ou bien

R_1 et R_2 forment ensemble un membre d'un pont choisi parmi -Y-CHR_{10}-CO-NR_{11}-, -O-CH_2-CO-NR_{12}-, -O-CH_2-CH_2-CZ- ou -CH=CH-NR_{13}-,

R_3 signifie un groupe C_{1-6}alkyle, C_{1-6}haloalkyle, C_{1-6}cyanoalkyle, C_{1-6}alcoxy, C_{3-6}cycloalcoxy, C_{3-6}alcényloxy, C_{3-6}alcynyloxy ou C_{1-6}alcoxy substitué par un groupe cyano, un halogène, C_{1-6}alcoxy, C_{1-6}alcoxycarbonyle, C_{3-6}alcynyloxycarbonyle ou bien
C_{1-4}alcoxy-C_{1-4}alcoxycarbonyle;

R_4 signifie l'hydrogène ou un groupe C_{1-4}alkyle;

R_5 signifie un groupe C_{1-4}alkyle, C_{3-4}alcényle, C_{3-4}alcynyle ou C_{1-4}alkyle substitué par un groupe cyano, formyle ou C_{1-4}alcoxycarbonyle;

R_6 signifie l'hydrogène ou un groupe C_{1-4}alkyle;

R_7 signifie un groupe C_{1-4}alkylsulfonyle, C_{1-4}alkyle ou C_{1-4}alkyle substitué par un groupe C_{1-4}alcoxycarbonyle;

R_8 signifie un groupe C_{1-4}alkyle;

R_9 signifie un groupe C_{1-4}alkyle, C_{1-4}cyanoalkyle, C_{1-4}alcoxy ou C_{3-4}alcényle; ou bien

R_8 et R_9 forment ensemble un pont C_{4-5}alkylène ou un pont éthylène-O-éthylène, ledit pont pouvant être substitué par un groupe C_{1-4}alkyle, ou bien
C_{1-4}alcoxycarbonyle;

R_{10} signifie l'hydrogène ou un groupe C_{1-4}alkyle;

R_{11} signifie l'hydrogène, un groupe C_{1-4}alkyle, C_{3-4}alcényle, C_{3-4}alcynyle ou C_{3-4}alcényle substitué par un halogène ou un groupe C_{1-4}alcoxycarbonyle;

R_{12} signifie un groupe C_{1-4}alcoxy;

R_{13} signifie un groupe C_{1-4}alkyle, C_{3-4}alcényle ou C_{3-4}alcynyle,

Y signifie l'oxygène ou le soufre, et

Z signifie l'oxygène ou un groupe =N-C_{1-4}alcoxy.

2. Un composé selon la revendication 1, où X signifie l'oxygène.

3. Un composé selon l'une quelconque des revendications 1 ou 2 où R signifie le fluor et R_1 signifie le chlore.

4. Un composé selon la revendication 1, où R_1 et R_2 forment ensemble le pont -O-CH_2-CO-NR_{11}-.

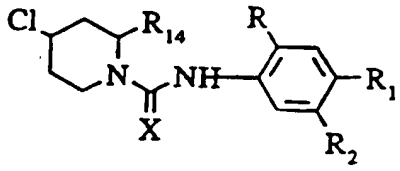
5. Un composé selon la revendication 1, où R_2 signifie un halogène, un groupe C_{1-6}alcoxycarbonyl-C_{1-4}alkyle ou C_{1-6}alcoxycarbonyl-C_{1-6}alcényle.

6. Un composé selon la revendication 1, où X signifie l'oxygène, R signifie le fluor, R_1 signifie le chlore et R_2 signifie un halogène, C_{1-6}alcoxycarbonyl-C_{1-4}alkyle ou C_{1-6}alcoxycarbonyl-C_{2-4}alcényle.

7. Un composé selon la revendication 1 choisi parmi le groupe comprenant la 7-chloro-2-(5-bromo-4-chloro-2-fluorophényl)-perhydroimidazo-[1,5a]pyridine-1,3-dione;

le β-[2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-phényl]-propionate de méthyle; et
le 2-chloro-4-fluoro-5-(7-chloro-perhydroimidazo[1,5a]pyridine-1,3-dione-2-yl)-cinnamate de méthyle.

8. Un procédé de préparation d'un composé de formule I selon la revendication 1, comprenant la condensation intramoléculaire d'un composé de formule II



(II)

où X, n, R, R_1 et R_2 sont tels que définis à la revendication 1, R_{1-4} signifie COOH, COOW et COSW, et W signifie un groupe C_{1-4}alkyle.

9. Une composition herbicide comprenant un composé de formule I selon la revendication 1 et un support acceptable en agriculture.

10. Une méthode de lutte contre les mauvaises herbes, qui comprend l'application aux mauvaises herbes ou à leur lieu de croissance d'une quantité efficace du point de vue herbicide d'un composé de formule I selon la revendication 1.